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(54) Power generating equipment comprising solid oxide fuel cells
Festoxidbrennstoffzellen enthaltendes Energieerzeugungsgerät

Dispositif générateur de courant électrique comportant des piles à combustible à oxydes solides

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#### Description

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The present invention relates to power generating equipment, particularly, relates to power generating equipment in which electric power is generated with the aid of solid oxide fuel cells.

Recently, fuel cells have been noted as a power generating source. The fuel cell is capable of directly converting chemical energy possessed by fuel to electric energy. Since the fuel cell is free from the limitation of Carnot's cycle, the fuel cell essentially has a high energy conversion efficiency. Further, various fuels such as naphtha, natural gas, methanol, coal reformed gas and heavy oil may be used, additionally, these fuels may be used with a low pollutant level. Moreover, the power generating efficiency of the fuel cells is not influenced by the scale of the equipment. Therefore, power generating equipment using solid oxide fuel cells is an extremely promising technique.

Particularly, since the solid oxide fuel cell (hereinafter abbreviated as SOFC) operates at a high temperature of 1000°C or more, the activity of electrodes thereof is very high and the use of a noble metal catalyst such as expensive platinum is not required. In addition, since the SOFC has a low polarization and a relatively high output voltage, the energy conversion efficiency is considerably higher than that of the other fuel cells. Furthermore, since the SOFC is constructed with solid materials, it is stable in structure and has a long life use.

Fig. 1 is a cross-sectional view showing an example of a conventional power generating equipment using hollow-cylindrical type SOFCs.

In Fig. 1, a round bottomed hollow-cylindrical SOFC element 5 comprising a round bottomed cylindrical porous support tube 6, an air electrode 7 formed on an outer periphery thereof, and a solid electrolyte 8 and a fuel electrode 9 arranged around the outer periphery of the air electrode 7 in this order. Generally, a plurality of such SOFC elements 5 are connected in series and parallel to constitute a collected cell; and the collected cell is provided at a given position in a power generating chamber 13. It, however, should be noted that only one SOFC element is drawn in Fig. 1 in order to make the explanation therefore easier. At the lower side of the power generating chamber 13, is provided a fuel gas chamber 14; and the fuel gas chamber 14 is separated from the power generating chamber 13 by a bottom side division wall 11. Additionally, at the lower side of the fuel gas chamber 14, is arranged a heat insulating wall 12. On the other hand, at the upper side of the power generating chamber 13, is arranged a gas exhaust chamber 3, which is separated from the power generating chamber 13 by an upper side division wall 4. In the upper side division wall 4, are provided a hole 4a, through which an opening side end portion of the SOFC element 5 is inserted. At the upper side of the gas exhaust chamber 3, is arranged a heat insulating wall 1, and an oxidizing gas supply tube 2 is inserted through a hole 1a formed in the wall 1 so as to be held by the through hole 1a. Then, a top end opening 2a of the oxidizing gas supply tube 2 is positioned in an internal space 10 of the SOFC element 5.

When operating the hollow-cylindrical SOFC, an oxidizing gas is supplied into the oxidizing gas supply tube 2 as shown by an arrow A, then the flow of the oxidizing gas supplied from the tube 2 is converted at the bottom portion of the element 5 and the gas is fed through the internal space 10 to be exhausted into the gas exhaust chamber 3 as shown by arrows B. On the other hand, when a fuel gas is supplied into a fuel gas chamber 14 through a fuel gas supply hole 12a formed in the heat insulating wall 12 as shown by an arrow C, pressure in the fuel gas chamber 14 becomes high; then the fuel gas is supplied into the power generating chamber 13 through the fuel gas supply holes 11a formed in the division wall 11, as shown by arrows D. And then the fuel gas is fed in an upper direction along the surface of the fuel electrode 9 of the SOFC element 5. Oxygen ion diffused in the solid electrolyte 8 is reacted with the fuel gas on the surface of the fuel electrode 9; as a result, an electric current is fed between the air electrode 7 and the fuel electrode 9. The used fuel gas is exhausted into the gas exhaust chamber 3 through the space 4a formed between the upper side division wall 4 and an opening end portion of the SOFC element 5, as shown by arrows E. The power generating equipment having no sealing as shown in Fig. 1 is preferred to be used, because the SOFC element 5 is operated under a high temperature about 1000°C.

In order to put such power generating equipment in a practical use, it is necessary to decrease the manufacturing cost therefore and to increase the power density of the equipment. Therefore, it is necessary to make the length of the SOFC element 5 long to increase the power generating output per one SOFC element.

However, in the power generating equipment having its constitution as shown in Fig. 1, there is a drawback that a temperature gradient is generated in the power generating chamber 13 due to the concentration gradient of the fuel gas flow in the chamber 13. That is to say, in the vicinity of the through holes 11a, through which the fuel gas is supplied into the chamber 13, the content of fuel is still large, so that a large amount of fuel is consumed by an electrochemical reaction there and hence the temperature of the atmosphere in the vicinity of the through holes 11a increases. Due to the temperature increase, the electrochemical reaction of the oxygen ion and the fuel on the fuel electrode 9 is activated more and more.

On the other hand, as far from the fuel supply holes 11a, the concentration of the fuel gas becomes low, and then the fuel amount consumed electrochemically is also decreased. Therefore, the temperature at the upper portion of the fuel electrode 9 is not increased so much and the electrochemical reaction is not activated so much. Additionally, a large amount of CO<sub>2</sub>, steam, etc. is contained in the fuel gas, whose concentration has been reduced, and the CO<sub>2</sub>

or steam adheres on the surface of the fuel electrode 9 to obstruct the reaction. Thus, the electrochemical reaction becomes more inactive and more in the upper side of the chamber 13.

Therefore, there is a large temperature gradient which is generated between the upperstream side and the downstream side of the fuel gas flow in the chamber 13. When the power generating equipment is operated for a long time, the temperature gradient not only causes cracks on the SOFC element 5 but also give a bad influence on the power generating efficiency thereof. This tendency becomes more considerable as each of cylindrical SOFC elements 5 is prolonged.

EP-A-0242200 proposes a fuel cell generator in which fuel gas is supplied via porous fuel feed conduits, which extend substantially the full length of the fuel cells. The fuel diffuses out of the conduits along the entire active length of the fuel cells. EP-A-0410796 describes a similar fuel cell generator in which the fuel feed conduits have openings spaced along their length. US 4983471 also proposes a system to distribute fuel along the length of fuel cells, in this case by providing a central fuel distribution pipe feeding transverse apertures.

The present invention has for its object to provide a novel power generating equipment in which the fuel concentration gradient in the fuel gas flow in the power generating chamber is small to reduce the temperature gradient in the chamber caused thereby.

The power generating equipment according to the invention is as set forth in claim 1.

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Since the power generating equipment according to the present invention comprises the fuel gas introducing means by which the fuel gas concentration in the power generating chamber becomes substantially even in a longitudinal direction of the solid oxide fuel cell elements, it is possible to prevent to generate a temperature gradient due to the concentration gradient of the fuel gas flow in the power generating chamber. Therefore, according to the invention, cracks on the solid oxide fuel cell elements are apt not to be generated and a power generating efficiency of the equipment can be improved.

In the power generating equipment according to the invention the fuel gas supply means comprises a partition, which constitutes at least a part of a side wall of said power generating chamber, preferably being arranged to be parallel with the longitudinal direction of the SOFC elements; the partition being for separating said fuel gas chamber from the power generating chamber and said fuel gas being supplied into the power generating chamber via the third partitions.

In the present invention, since the fuel gas is supplied into the power generating chamber via the partition, the gas concentration of the fuel gas flow in the power generating chamber becomes substantially even in the power generating chamber in the longitudinal direction of the SOFC elements.

It should be noted that in the present invention the fuel gas means a gas including hydrogen, reformed hydrogen, carbon monoxide, etc; and the oxidizing gas means a gas including an oxidizing agent such as oxide and hydrogen peroxide.

Fig. 1 is a partial cross-sectional view showing a construction of a conventional power generating equipment;

Fig. 2 is a cross-sectional view depicting a construction of a first embodiment of a power generating equipment according to the invention;

Fig. 3 is a partial plan view illustrating a construction of the first embodiment depicted in Fig. 2;

Fig. 4 is an enlarged-scale plan view indicating a construction of the first embodiment illustrated in Fig. 3; and

Fig. 5 is a cross-sectional view representing a construction of a second embodiment of a power generating equipment according to the invention.

Fig. 6, 7 and 8 show the construction of a power generating equipment described for comparison with the present invention.

Fig. 6 is a cross-sectional view showing a construction of a power generating equipment described for comparison with the invention viewed from a direction perpendicular to the longitudinal direction of SOFC elements; Fig. 7 is a cross-sectional view depicting a construction of the equipment viewed from a direction parallel to the longitudinal direction of SOFC elements; and Fig. 8 is a partial enlarged-scale view of the construction shown in Fig. 6.

A plurality of bottomed cylindrical SOFC elements 28 are connected to each other in series and parallel to constitute a collecting cell. In Fig. 6, four elements 28 are connected to each other in series in a vertical direction and three rows of the elements 28 are connected in parallel in a horizontal direction. It should be noted that the numbers of cells and rows may be changed.

As clearly shown in Fig. 8, each SOFC element 28 comprises a porous ceramic tube 36; and on an outer circumference of the tube 36 are arranged an air electrode 37, solid electrolyte 38 and fuel electrode 39, successively. Further, the solid electrolyte 38 and the fuel electrode. 39 are not provided all around the outer surface of the air electrode 37; and in a rift of the solid electrolyte 38 and the fuel electrode 39 is arranged an interconnector 29 and a connecting terminal 30 from an inner side in this order. The SOFC elements 28 contiguously arranged in a vertical direction in Fig. 8 are connected in series to each other in such a manner that the air electrodes 37 are connected to the fuel electrodes

39 of the contiguous SOFC elements 28 via the interconnector 29 and the connecting terminal 30 and a metal felt 31.

For instance, four SOFC elements 28 are connected to each other in series as showing in Fig. 6. On the other hand, between the SOFC elements 28 contiguously arranged in a horizontal direction in Figs. 6 and 8, is arranged a fuel gas supply tube 26. In this comparative example, two fuel gas supply tubes 26 are arranged in the respective space formed between the SOFC elements arranged in a horizontal direction, and the other fuel gas tubes 26 which are provided at both sides of the rows of the element 28.

The SOFC elements 28 are connected to each other in series and parallel in such a manner as to constitute a collecting cell as shown in Fig. 6. It, however, should be noted that in Figs. 6 and 7, the detail construction of each SOFC element is not indicated and that the interconnector 29 and the connecting terminal 30 are depicted as a single part as a matter of convenience.

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The positive and negative electrodes of the thus constructed collecting cell are electrically connected to current collecting boards 24 via the metal felt 31, respectively. The pair of current collecting boards 24 serve as a current collecting member of the collecting cell for collecting an electric power generated in each SOFC element 28; and are connected to lead lines (not shown). The collecting cell and the collecting boards 24 are contained in a power generating chamber 25 as a single body.

The air electrode 37 can be made by doped or non-doped conductive perovskite-type oxide, such as LaMnO<sub>3</sub>, CaMnO<sub>3</sub>, LaNiO<sub>3</sub>, LaCoO<sub>3</sub> and LaCrO<sub>3</sub>; but particularly, LaMnO<sub>3</sub>, in which strontium is doped, is preferably used as a material of the air electrode 37. The solid electrolyte 38 may be preferably made by yttria stabilized zirconia or yttria partially stabilized zirconia. As a material of the fuel electrode 39, nickel-zirconia cermet or cobalt zirconia cermet is preferred. Additionally, the interconnecter 29 may be preferably made by doped or non-doped perovskite-type LaCrO<sub>3</sub> or LaMnO<sub>3</sub>.

The fuel gas supply tube 26 is made of a porous conductive material or a porous non-conductive material. As a porous non-conductive material, the non-conductive ceramics such as porous zirconia and alumina is preferred; and as a porous conductive material, a porous metal made by sintering a reduction resistance metal or a porous cermet made by sintering a mixture of the reduction resistance metal powder and a ceramic powder is preferred.

As the ceramic powder, for instance, a ceramic powder mainly composing alumina or zirconia can be indicated. And, as the reduction resistance metal powder, an alloy powder such as Ni-Cr, Ni-Fe-Cr, Ni-Fe-Cr-Al, Co-Ni-Cr, Fe-Cr and Fe-Cr-Al, or a powder of Ni, Co or Fe can be shown as an example.

It is preferred that the porosity of the fuel gas supply tube 26 is about 10~90%. When the porosity exceeds over 90%, the strength of the tube 26 would be decreased; and when the porosity is less than 10%, the fuel gas would not be apt to be run through the tube 26.

It may be possible to give a gradient on the porosity of the fuel gas supply tube 26 so as to distribute the fuel gas in an even manner in the fuel gas chamber 25.

There are two ways to make a gradient on the porosity of the fuel gas supply tube 26 as in the following.

- (1) When sintering the fuel gas supply tube 26 made by a porous metal, porous cermet or porous ceramics, holding one end portion of the green body of the fuel gas supply tube and attaching a weight on the other end portion to hang the green body. By this hanging, a comparatively large load is given to the holding side portion of the green body to make the porosity thereof large; and only small load is given to the weighting side of the green body, then the porosity thereof becomes comparatively small.
- (2) Tubular-shaped sintered body made by a porous metal, porous cermet or porous ceramics is prepared; then a filler material is impregnated in the pores of the sintered body to fill the pores to some degree with the material; then the sintered body is dried or heated to remain the filler material in the pores. By varying the impregnating amount of the filler material, it is possible to make a gradient in the porosity of the sintered body.

The following is a method for controlling the fuel gas amount blown from the outer circumference surface of the tube 26 into the power generating chamber 25.

That is to say, the fuel gas supply tube is constructed in a double manner. Concretely, tubular sintered body made of porous metal is prepared; the porosity of the sintered body is arranged to be even; then a plurality of slurry bands are formed on the outer surface of the sintered body at a given distance; and then the body is sintered again. In this case, by making the width of slurry band large or by making the distance between the bands small, it is possible to control the transmission amount of the fuel gas through the tube 26 to be small. Contrary, by making the width of slurry band small or by making the distance between the bands large, the transmission of the fuel gas is accelerated. Further, by selecting the grain diameter of the reduction resistance metal powder or the ceramics powder included in the slurry, it is possible to control the gas transmission amount. In this case, it is preferred that the tubular sintered body is used as a back up material and the side thereof, on which the slurry is arranged, is faced to the power generating chamber.

Heat insulating densified material or reduction resistance densified material may be used as the material of the fuel gas supply tube 26. In this case, a plurality of through holes should be formed in the wall of the fuel gas supply

tube 26. The diameter, position and number of the through holes are determined so as to evenly distribute the fuel gas along the surface of the SOFC element 28. Nickel-series or Co-series alloys such as Hastelloy and Inconel (Trademark) are preferred to be used as a densified material of the fuel gas supply 26.

In the comparative example, each SOFC element 28 is held in a vertical direction, that is to say, the longitudinal direction of the SOFC element 28 is extended in the vertical direction. However, it may be possible to arrange that the longitudinal direction of the element is extended in the horizontal direction. Further, in the comparative example, round bottomed-cylindrical-shaped SOFC elements are used, but it may be possible to use cylindrical SOFC element having openings at both ends thereof.

Fig. 2 is a cross-sectional view showing a power generating equipment of first embodiment of the present invention; and Fig. 3 is a partial plan view representing one part of the equipment shown in Fig. 2. Further, Fig. 4 is a partial plan view indicating one part of the equipment shown in Fig. 3 in an enlarged-scale.

Round bottomed cylindrical SOFC elements 76 are connected in series and parallel to constitute a collecting cell. In the first embodiment, the SOFC elements 76 are arranged in four by four; and in Fig. 3, the longitudinally arranged elements are connected to each other in series and horizontally arranged elements are connected to each other in parallel, but this arrangement is possible to change as desired.

The construction of each SOFC element 76 is the same as that of the comparative example and connection thereof is also the same as that of the comparative example. In the first embodiment, the numerical number 87 represents a porous support tube, 88 air electrode, 89 solid electrolyte, 90 a fuel electrode; 86 an interconnector, 83 connecting terminal and 79 represents a metal felt.

The SOFC elements 76 are connected to each other in series and parallel to constitute a collective cell shown in Fig. 3, in which four by four SOFC elements are arranged. It should be noted that in Figs. 2 and 3, a detail construction of each SOFC element is not drawn and the interconnector 86 and the connecting terminal 83 are shown as a single body.

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The positive and negative electrodes of the thus structured collective cell are electrically connected to a pair of current collection boards 75 via the metal felt 79. The pair of current collection boards 75 serve as a current collector of the collective cell, and the boards 75 are connected to lead lines (not shown), respectively. The collective cell and the current collection boards 75 are contained in the power generating chamber 82 as a single body.

The power generating chamber 82 comprises a first wall 77 arranged at an opening side of SOFC element 76; in the first wall are provided circular through holes 77a so as to correspond to the SOFC elements 76. The SOFC elements 76 are inserted into the through holes 77a, respectively, to be held thereby. In the inner space of each SOFC element is inserted an oxidizing gas supply tube 80. The oxidizing gas is fed in an upper direction in Fig. 2 to be exhausted to the gas exhaust chamber 78. The gas flow is shown by arrows  $D_4$  in Fig. 2.

The power generating chamber 82 further comprises a second wall, which is arranged at a bottom side of SOFC element 76 so as to be perpendicular to the longitudinal direction of the SOFC element. On the second wall, are held the bottoms of the SOFC elements 76. The power generating chamber 82 further comprises a pair of side walls 74 arranged to be parallel to each other and also parallel to the longitudinal direction of the SOFC element 76. The side walls 74 are arranged to be separated from the current collection boards 75 at a some distance. A can-like body 71 made by a heat insulating material is arranged surrounding the power generating chamber 82.

Between the can-like body 71 and the second wall 73 and the side walls 74 is provided the fuel gas chamber 72; the cross-sectional view of the fuel gas chamber 72 is U-shaped as shown in Fig. 2; and the upper end of the fuel gas chamber 72 is separated from the exhaust gas chamber 78 by means of the upper wall of the can-like body 71. It should be noted that in the bottom of the can-like body 71, is provided a fuel gas supply hole 71a.

In the second wall 73 of the power generating chamber 82, a plurality of through holes for transmitting the fuel gas are regularly provided with a given distance. Further, in the respective side walls 74, there are provided a plurality of through holes 74a in a matrix manner. Furthermore, in the respective current collection boards 75 are provided a plurality of through holes 75a in a matrix manner so as to make the diameters and positions of the holes 74a in the side walls 74 coincident with those of the holes 75a in the current collection boards 75.

When operating the power generating equipment according to the fourth embodiment, the fuel gas is supplied into the fuel gas chamber 72 via the hole 71a as shown by an arrow  $A_4$ . Then, the pressure in the chamber 72 is increased and the fuel gas is supplied into the power generating chamber 82 via the through holes 73a arranged in the second wall 73 as shown by arrows B4; additionally, the fuel gas is also supplied into the power generating chamber 82 via the through holes 74a in the side walls 74 and through holes 75b in the current collection boards 75 as shown by arrows  $C_4$ . The thus supplied fuel gas is mainly fed in a horizontal direction in the chamber 82 as shown by arrbws  $C_4$  in Fig. 3. After electric power is generated in the chamber 82, the depleted fuel gas is finally exhausted out through the space between the SOFC elements 76 and the first wall 77 into the exhaust gas chamber 78; and then the depleted fuel gas is mixed with the depleted oxidizing gas therein.

The preferred materials for the air electrodes 88, solid electrolytes 89, fuel electrodes 90 and the interconnector 86 are the same as those cited in the comparative example.

The side walls 74 are made of a heat insulating metal or ceramics, which has a proof resistance against the fuel gas and is stable at an operating temperature of the SOFC elements. Alloys such as Ni-Cr, Ni-Fe-Cr, Ni-Fe-Cr-Al, Co-Ni-Cr, Fe-Cr and Fe-Cr-Al can be cited, for instance, as such heat insulating metals.

According to the first embodiment, since the equipment is arranged such that the fuel gas is supplied not only from the second wall of the chamber 82 but also the side walls of so-called cell unit as shown in Fig. 3, fresh fuel gas can be constantly supplied to all the surface of the SOFC element. Therefore, the fuel concentration gradient becomes small in the chamber 82 and substantially even, so that the temperature gradient along the longitudinal direction of the SOFC element 76 also becomes small. As a result, even when the equipment is operated for a long time, defects such as cracks are not apt to be generated in the SOFC element 76. Further, since the unevenness of the electrochemical reaction becomes small, it is possible to improve the power generating efficiency of the SOFC elements 76.

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Moreover, in the conventional equipment shown in Fig. 1, the fuel gas chamber is arranged only under the second wall, and therefore, the side walls of the power generating chamber are surrounded by, for instance, air. Additionally, since the temperature of the air is low, it is necessary to give a heat insulation to the side wall 74. In other words, the side walls should be designed as a heat insulating walls. Therefore, the thickness of the side walls should be made large, for example, several tens of millimeters.

Contrary to this, in the first embodiment according to the present invention, in the fuel gas chamber 72 surrounding the power generating chamber 82 is filled with the fuel gas having a high temperature. Therefore, even if the thickness of the side walls 74 is small, it is possible to keep the temperature in the power generating chamber 82 at the operating temperature of SOFC element 76. As a result, it is possible to make the size of the equipment compact.

In the first embodiment, the direction of the gas flow blown out from the side wall 74 is important. In the up and down directions in Fig. 3, in other words, the in series connected direction of SOFC elements 76, belt-shaped interconnectors are arranged along the longitudinal direction of the element 76, and corresponding to this, the metal felts 79 are arranged between the SOFC elements to fill the space thereby. Therefore, the fuel gas could not pass through the portion well. On the other hand, the metal felts 84, by which the SOFC elements 76 are connected to each other in parallel, are not necessary to have belt-like shapes, but the metal felts 84 may be arranged to connect the elements at only one or two portions between the SOFC elements.

Therefore, the fuel gas can pass through well in the up and low directions in Fig. 3; so that when the fuel gas is supplied into the chamber 82 in directions shown by arrows  $C_4$  in Fig. 3, the gas can be easily distributed all over the power generating chamber 82.

In the first embodiment shown in Fig. 2, in order to make the fuel gas concentration more even in the longitudinal direction of the SOFC element, the diameters and numbers of the through holes 74a and 75a formed in the side walls 74 and current correction boards 75 should be determined in a suitable manner.

Fig. 5 is a cross sectional view showing the construction according to a second embodiment of the present invention.

In the second embodiment, both the second wall 93 and a pair of side walls 94 are made of a porous material. Therefore, the fuel gas in the fuel gas chamber 72 is transmitted through the first wall 93 (arrows  $B_5$ ) and the side walls 94 (arrows  $C_5$ ) to be fed into the power generating chamber 82 by a differential pressure between the gas exhaust chamber 78 and the fuel gas chamber 72. The fuel gas transmitted through the side walls 75 further passes through the through holes 75a and then flows along with the surface of the fuel electrodes of the SOFC elements 76. Therefore, in the second embodiment, it is possible to obtain the same effect as the first embodiment. Additionally, in the second embodiment, since the side walls as a whole are made of the porous material, it would be possible to distribute the fuel gas in the chamber 82 in a more even manner.

However, if the porosity of the side walls is even, the gas concentration in the vicinity of the bottom side of the element 76 would become higher than that in the vicinity of the opening side of the element 76. Therefore, it is desired that the porosity of the side walls has a gradient so as to the transmitted gas amount in the upper side becomes larger than that in the bottom side.

Further, the first wall 93 and the side walls 94 should be stabilized against the high temperature fuel gas. Therefore, the porous metal made by sintering the reduction resistance metal powder or the porous cermet made by sintering the reduction resistance metal powder and ceramic powder, is preferred as the material thereof. The ceramic powder mainly consisting of alumina or zirconia is cited as the ceramic powder, and alloy powder such as Ni-Cr, Ni-Fe-Cr, Ni-Fe-Cr-Al, Co-Ni-Cr, Fe-Cr and Fe-Cr-Al or metal powder such as Ni, Co and Fe are cited as the reduction resistant metal powder.

It is preferred that the side wall 94 has its porosity of 10~90%, as in the comparative example. It is further preferred to make a gradient in the porosity of the side wall 94; and the gradient can be made in the same manner explained in the comparative example. Additionally, fuel gas transmitting amount control, with the aid of slurry, can be conducted in the same manner as for the fuel gas supply tube in the comparative example.

Each SOFC element is held in a vertical direction. However, it may be possible to hold the SOFC element in a horizontal direction.

In the first and second embodiments, since the fuel gas with a high temperature is supplied from outside of the side walls into the power generating chamber, it is not necessary to give a heat insulating function to the side walls. Therefore, even if the thickness of the side wall is thin, the temperature in the power generating chamber can be kept at an operational temperature of the SOFC elements; and thus it is possible to make the size of the power generating equipment compact.

The present inventors conducted an experiment to measure the temperature gradients in the power generating chambers of the conventional equipment shown in Fig. 1 and the present equipment shown in Fig. 2 in the longitudinal direction of the SOFC elements provided therein. The measuring points a, b, c, d and e indicated in Fig. 2 were selected, and the distance between the respective measuring points is 100mm. A gas containing 96% of hydrogen and 4% of steam is used as the fuel gas, and air is used as the oxidizing gas. The temperature of the SOFC element is measured with the aid of thermo couples. Further, a power output per one SOFC element of each equipment is also measured. The measurement result is shown in Table 1.

Table 1

Table I						
	Temperature (°C)					Output (W)
	а	ь	С	d	е	
Conventional equipment (Fig. 1)	1024	1020	978	901	833	6.6
Fourth embodiment (Fig. 7)	1037	1033	1001	972	964	8.1

As is clear from Table 1, since the power generating equipment according to the present invention is constructed such that the fuel gas is supplied into the power generating chamber in an even manner in a longitudinal direction of SOFC elements provided in the chamber, a fresh fuel gas can be constantly supplied to not only bottom side of the SOFC element but also to the upper opening side thereof. Therefore, the fuel gas concentration gradient in the power generating chamber becomes small and the concentration becomes even therein; additionally the temperature gradient in the longitudinal direction of SOFC element in the chamber becomes small too. As a result, even when the power generating equipment is operated for a long time, cracks are not apt to be generate in the SOFC elements. Moreover, an evenness of the electrochemical reaction becomes small, so that the power generating efficient of each SOFCE element is improved.

#### Claims

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A power generating apparatus comprising:

a plurality of cylindrical solid oxide fuel cell elements (76) electrically connected to each other to constitute a collective cell, said fuel cell elements (76) extending in a longitudinal direction of said cell; current collecting means (75) connected to positive and negative electrodes of said collective cell;

a power generating chamber (82) containing said collective cell and said current collecting means;

a fuel gas chamber (72) separated from said power generating chamber (82) by means of a partition (74,76; 93,94);

an oxidizing gas supply means (80) for supplying an oxidizing gas into an internal space of each of said solid oxide fuel cell elements (76) and a fuel gas supply means (74,74a;94) for supplying a fuel gas from said fuel gas chamber (72) to said power generating chamber through said partition (74,76;93,94) whereby the oxidizing gas is electrochemically reacted with the fuel gas to generate electric power, said fuel gas supply means (74,74a;94) introducing said fuel gas into said power generating chamber (82) in a manner such that the fuel gas concentration in said power generating chamber is substantially uniform in the longitudinal direction of said solid oxide fuel cell elements (76);

characterised in that said fuel gas supply means comprises, as at least part of said partition, at least one porous or apertured first side wall (74;94) bounding said power generating chamber (82) at the exterior periphery of the power generating chamber and extending in said longitudinal direction, the fuel gas passing through the pores or apertures (74a) of said first side wall or walls (74;94) inwardly towards the power generating chamber from said fuel gas chamber (72).

A power generating apparatus according to claim 1, wherein each said solid oxide fuel cell element (76) comprises
a porous supporting tube (87) and an air electrode (88), solid electrolyte (89) and fuel electrode (90) arranged

around an outer surface of said porous supporting tube (87) in this order.

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- 3. A power generating apparatus according to claim 2, wherein said air electrode (88) of each solid oxide fuel cell element is electrically connected in series to a fuel electrode (90) of another solid oxide fuel cell element which is contiguously arranged in a first direction.
- 4. A power generating apparatus according to claim 2 or claim 3, wherein said air electrode (88) of each solid oxide fuel cell element is connected to the fuel electrode (90) of a contiguously arranged solid oxide fuel cell via an interconnector (86), a connecting terminal (83) and a metal felt (79).
- A power generating apparatus according to claim 4, wherein each said interconnector (86) is made of doped or non-doped conductive perovskite-type oxide of LaCrO<sub>3</sub>.
- 6. A power generating apparatus according to claim 3, wherein a metal felt (84) is arranged between each solid oxide fuel cell element (76) in a second direction perpendicular to said first direction, and said solid oxide fuel cell elements (76) contiguously arranged in said second direction are electrically connected to each other in parallel by said metal felts (84).
- A power generating apparatus according to any one of claims 5 to 6, wherein said air electrode is made of doped or non-doped conductive perovskite type oxide such as LaMnO<sub>3</sub>, CaMnO<sub>3</sub>, LaNiO<sub>3</sub>, LaCoO<sub>3</sub> or LaCrO<sub>3</sub>.
  - 8. A power generating apparatus according to any one of claims 2 to 7, wherein said solid electrolyte (89) is made of yttria stabilized zirconia or yttria partially stabilized zirconia.
- A power generating apparatus according to any one of claims 2 to 8, wherein said fuel electrode (90) is made of nickel-zirconia cermet or cobalt-zirconia cermet.
  - 10. A power generating apparatus according to any one of claims 1 to 9, wherein said fuel gas supply means further comprises, as part of said partition, a second wall (73;93) by which said power generating chamber (82) is separated from said fuel gas chamber (72), arranged in a direction perpendicular to said longitudinal direction.
  - 11. A power generating apparatus according to any one of claims 1 to 10, wherein said current collecting means (75) comprises a pair of current collecting boards and the boards are arranged between said collective cell and said first side walls (74;94) of said power generating chamber, parallel to said longitudinal direction.
  - 12. A power generating apparatus according to claim 13, wherein a pturality of through holes (75a) are provided in said current collecting boards (75).
  - 13. A power generating apparatus according to any one of claims 1 to 12, wherein a plurality of through holes (73a, 74a) through which the fuel gas passes into said power generating chamber, are provided in at least said first walls (74).
    - 14. A power generating apparatus according to any one of claims 1 to 12, wherein at least said first walls (93,94) are made of porous material.
    - 15. A power generating apparatus according to claim 14, wherein at least said first walls (74) are made of porous metal formed by sintering a reduction-resistant metal powder or are made of a porous cermet formed by sintering a mixture of a reduction-resistant metal powder and a ceramic powder.
- 50 16. A power generating apparatus according to claim 15, wherein at least said first walls (74) are made of nickel-zirconia cermet or cobalt-zirconia cermet.
  - 17. A power generating apparatus according to any one of claims 14 to 16, wherein at least said first walls (74) have a porosity in the range 10 to 90%.
  - 18. A power generating apparatus according to claim 17, wherein at least said first walls (74) each comprise a sintered body made of porous metal having a given porosity and a plurality of belt-shaped regions formed of slurries arranged on the surface of the sintered body at predetermined spacings.

19. A power generating equipment according to claim 18, wherein the amount of the fuel gas transmitted through said first walls (74) is controlled by varying said spacings between the slurries.

#### 5 Patentansprüche

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1. Energierzeugungsvorrichtung bzw. -apparat bzw. -gerät, umfassend:

eine Vielzahl zylindrischer Festoxidbrennstoffzellenelemente (76), die elektrisch miteinander verbunden sind, um eine Sammel- bzw. Mehrfachzelle zu bilden, wobei sich die Brennstoffzellenelemente (76) in einer Längsrichtung der Zelle erstrecken;

Stromsammelmittel (75), die mit positiven und negativen Elektroden der Sammelzelle verbunden sind; eine Energieerzeugungskammer (82), welche die Sammelzelle und die Stromsammelmittel enthält; eine Brenngaskammer (72), die mittels einer Trennwand (74, 76; 93, 94) von der Energieerzeugungskammer getrennt ist;

ein Zufuhrmittel (80) für oxidierendes Gas zum Zuführen eines oxidierenden Gases in einen Innenraum jedes der Festoxidbrennstoffzellenelemente (76) und ein Brenngaszufuhrmittel (74, 74a; 94) zum Zuführen eines Brenngases von der Brenngaskammer (72) in die Energieerzeugungskammer durch die Trennwand (74, 76; 93, 94) hindurch, wodurch das oxidierende Gas elektrochemisch mit dem Brenngas umgesetzt bzw. reagieren gelassen wird, um elektrische Energie zu erzeugen, wobei das Brenngaszufuhrmittel (74, 74a; 94) das Brenngas solcherart in die Energieerzeugungskammer (82) leitet, daß die Brenngaskonzentration in der Energieerzeugungskammer in Längsrichtung der Festoxidbrennstoffzellenelemente (76) im wesentlichen einheitlich ist;

dadurch gekennzeichnet, daß das Brenngaszufuhrmittet als zumindest ein Teil der Trennwand zumindest eine poröse oder mit Öffnungen versehene erste Seitenwand (74; 94) umfaßt bzw. aufweist, welche die Energieerzeugungskammer (82) am Außenumfang der Energieerzeugungskammer begrenzt und sich in Längsrichtung erstreckt, wobei das Brenngas durch die Poren oder Öffnungen (74a) der ersten Seitenwand oder Seitenwände (74; 94) hindurch von der Brenngaskammer (72) nach innen zur Energieerzeugungskammer hin strömt.

- Energieerzeugungsvorrichtung nach Anspruch 1, worin jedes Festoxidbrennstoffzellenelement (76) ein poröses Stützrohr (87) und eine Luftelektrode (88), einen Trockenelektrolyt (89) und eine Brennstoffelektrode (90) in dieser Reihenfolge um eine Außenfläche des porösen Stützrohrs (87) herum angeordnet umfaßt.
- 35 3. Energieerzeugungsvorrichtung nach Anspruch 2, worin die Luftelektrode (88) jedes Festoxidbrennstoffzellenelements mit einer Brennstoffelektrode (90) eines anderen Festoxidbrennstoffzellenelements, das in einer ersten Richtung daran angrenzend angeordnet ist, elektrisch in Serie verbunden ist.
- 4. Energieerzeugungsvorrichtung nach Anspruch 2 oder 3, worin die Luftelektrode (88) jedes Festoxidbrennstoffzel40 lenelements mit der Brennstoffelektrode (90) einer daran angrenzend angeordneten Festoxidbrennstoffzelle über
  einen Zwischenverbinder (86), einen Verbindungsanschluß (83) und einen Metallfilz (79) verbunden ist.
  - 5. Energieerzeugungsvorrichtung nach Anspruch 4, worin jeder Zwischenverbinder (86) aus dem dotierten oder nichtdotierten, leitenden, perowskitartigen Oxid LaCrO<sub>3</sub> besteht.
  - Energieerzeugungsvorrichtung nach Anspruch 3, worin ein Metallfilz (84) zwischen jedem Festoxidbrennstoffzellenelement (76) in einer zweiten Richtung senkrecht zur ersten Richtung angeordnet ist und die in der zweiten Richtung aneinander angrenzend angeordneten Festoxidbrennstoffzellenelemente (76) durch die Metallfilze (84) parallel miteinander elektrisch verbunden sind.
  - Energieerzeugungsvorrichtung nach einem der Ansprüche 5 oder 6, worin die Luftelektrode aus dotiertem oder nichtdotiertem, leitendem, perowskitartigem Oxid, wie z.B. LaMnO<sub>3</sub>, CaMnO<sub>3</sub>, LaNiO<sub>3</sub>, LaCoO<sub>3</sub> oder LaCrO<sub>3</sub>, besteht.
- Energieerzeugungsvorrichtung nach einem der Ansprüche 2 bis 7, worin der Festelektrolyt (89) aus mit Yttriumoxid stabilisiertem Zirkonoxid oder aus teilweise mit Yttriumoxid stabilisiertem Zirkonoxid besteht.
  - 9. Energieerzeugungsvorrichtung nach einem der Ansprüche 2 bis 8, worin die Brennstoffelektrode (90) aus Nickel-

Zirkonoxid-Cermet oder Kobalt-Zirkonoxid-Cermet besteht.

- 10. Energieerzeugungsvorrichtung nach einem der Ansprüche 1 bis 9, worin das Brenngaszufuhrmittel weiters als Teil der Trennwand eine zweite Wand (73; 93) umfaßt, durch welche die Energieerzeugungskammer (82) von der Brenngaskammer (72) getrennt ist, die in einer zur Längsrichtung senkrechten Richtung angeordnet ist.
- 11. Energieerzeugungsvorrichtung nach einem der Ansprüche 1 bis 10, worin das Stromsammelmittel (75) ein Paar Stromsammelplatten umfaßt und die Platten zwischen der Sammelzelle und den ersten Seitenwänden (74; 94) der Energieerzeugungskammer parallel zur Längsrichtung angeordnet sind.
- 12. Energieerzeugungsvorrichtung nach Anspruch 11, worin eine Vielzahl an Durchgangslöchern (75a) in den Stromsammelplatten (75) ausgebildet ist.
- Energieerzeugungsvorrichtung nach einem der Ansprüche 1 bis 12, worin eine Vielzahl an Durchgangslöchern
   (73a, 74a), durch die das Brenngas in die Energieerzeugungskammer gelangt, in zumindest den ersten Wänden
   (74) ausgebildet ist.
  - Energieerzeugungsvorrichtung nach einem der Ansprüche 1 bis 12, worin zumindest die ersten Wände (93, 94) aus porösem Material bestehen.
  - 15. Energieerzeugungsvorrichtung nach Anspruch 14, worin zumindest die ersten Wände (74) aus porösem Metall, das durch Sintern eines reduktionsbeständigen Metallpulvers gebildet ist, oder aus einem porösen Cermet bestehen, das durch Sintern eines Gemischs aus einem reduktionsbeständigen Metallpulver und einem Keramikpulver gebildet ist.
  - Energieerzeugungsvorrichtung nach Anspruch 15, worin zumindest die ersten W\u00e4nde (74) aus Nickel-Zirkonoxid-Cermet oder Kobalt-Zirkonoxid-Cermet bestehen.
  - Energieerzeugungsvorrichtung nach einem der Ansprüche 14 bis 16, worin zumindest die ersten Wände (74) eine Porosität im Bereich von 10-90% aufweisen.
    - 18. Energieerzeugungsvorrichtung nach Anspruch 17, worin zumindest die ersten Wände (74) jeweils einen Sinterkörper aus porösem Metall mit einer bestimmten Porosität und einer Vielzahl bandförmiger Bereiche umfassen, die aus Aufschlämmungen gebildet sind, die in vorbestimmten Abständen auf der Oberfläche des Sinterkörpers ausgebildet sind.
    - Energieerzeugungsvorrichtung nach Anspruch 18, worin die Menge des durch die ersten W\u00e4nde (74) str\u00f6menden Brenngases durch Variieren der Abst\u00e4nde zwischen den Aufschl\u00e4mmungen gesteuert wird.

#### Revendications

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- 1. Dispositif de production d'énergie comprenant :
- un certain nombre d'éléments formant des piles à combustible à oxydes solides (76) reliés électriquement les uns aux autres pour constituer une pile de collecte, lesdits éléments à piles à combustible (76) s'étendant dans une direction longitudinale de ladite pile;
  - un moyen de collecte de courant (75) relié à des électrodes positive et négative de ladite pile de collecte;
  - une chambre de production d'énergie (82) contenant ladite pile de collecte et ledit moyen de collecte de courant;
- une chambre de gaz de combustible (72) séparée de ladite chambre de production d'énergie (82) au moyen d'une séparation (74, 76 ; 93, 94) ;
  - un moyen d'alimentation en gaz d'oxydation (80) pour fournir un gaz d'oxydation dans un espace interne de chacun desdits éléments à pile à combustible à oxydes solides (76) et un moyen d'alimentation en gaz de

combustible (74, 74a; 94) pour fournir un gaz de combustible de ladite chambre de gaz de combustible (72) à ladite chambre de production d'énergie à travers ladite séparation (74, 76; 93, 94) de la sorte le gaz d'oxydation est électrochimiquement réagi avec le gaz de combustible pour produire de l'énergie électrique, ledit moyen d'alimentation en gaz de combustible (74, 74a; 94) introduisant ledit gaz de combustible dans ladite chambre de production d'énergie (82) d'une manière telle que la concentration de gaz de combustible dans ladite chambre de production d'énergie est sensiblement uniforme dans la direction longitudinale desdits éléments à cellules à combustible à oxydes solides (76);

caractérisé en ce que ledit moyen d'alimentation en gaz de combustible comprend, comme au moins une partie de ladite séparation, au moins une première paroi latérale poreuse ou ajourée (74; 94) liant ladite chambre de production d'énergie (82) à la périphérie extérieure de la chambre de production d'énergie et s'étendant dans ladite direction longitudinale, le gaz de combustible passant à travers les pores ou les ouvertures 74a) de ladite première paroi latérale ou des parois (74; 94) intérieurement vers la chambre de production d'énergie à partir de la chambre de gaz de combustible (72).

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dans cet ordre.

La chambre de gaz de combustible (72).
 Dispositif de production d'énergie selon la revendication 1, dans lequel chaque élément à cellule à combustible à oxyde solide précité (76) comprend un tube de support poreux (87) et une électrode d'air (88), un électrolyte solide (89) et une électrode de combustible (90) agencés autour de la surface externe dudit tube de support poreux (87)

3. Dispositif de production d'énergie selon la revendication 2, dans lequel l'électrode d'air précitée (88) de chaque élément à pile à combustible à oxyde solide est électriquement reliée en série à une électrode de combustible (90) d'un autre élément à pile à combustible à oxyde solide qui est agencé de façon contigue dans une première direction.

4. Dispositif de production d'énergie selon la revendication 2 ou la revendication 3, dans lequel l'électrode d'air précitée (88) de chaque élément à pile à combustible à oxyde solide est reliée à l'électrode de combustible (90) d'une pile à combustible à oxyde solide agencée de façon continue par l'intermédiaire d'un interconnecteur (86), d'une borne de connexion (83) et d'un feutre métallique (79).

 Dispositif de production d'énergie selon la revendication 4, dans lequel chaque interconnecteur précité (86) est réalisé en oxyde de LacrO<sub>3</sub> de type perovskite conducteur dopé ou non dopé.

- 6. Dispositif de production d'énergie selon la revendication 3, dans lequel un feutre métallique (84) est agencé entre chaque élément à pile à combustible à oxyde solide (76) dans une seconde direction perpendiculaire à la première direction précitée et les éléments à pile à combustible à oxydes solides précités (76) agencés de façon contiguë dans ladite seconde direction sont reliés électriquement les uns aux autres en parallèle par lesdits feutres métalliques (84).
- 7. Dispositif de production d'énergie selon l'une quelconque des revendications 5 à 6, dans lequel l'électrode d'air précitée est réalisée en oxyde de type perovskite conducteur dopé ou non dopé tel que LaMnO<sub>3</sub>, CaMnO<sub>3</sub>, LaNiO<sub>3</sub>, LaCoO<sub>3</sub> ou LaCrO<sub>3</sub>.
- Dispositif de production d'énergie selon l'une quelconque des revendications 2 à 7, dans lequel l'électrolyte solide précité (89) est réalisé en zircone stabilisée par de l'oxyde d'yttrium ou zircone stabilisée partiellement par de l'oxyde d'yttrium.
  - Dispositif de production d'énergie selon l'une quelconque des revendications 2 à 8, dans lequel l'électrode de combustible précitée (90) est réalisée en cermet nickel-zircone ou cobalt-zircone.
  - 10. Dispositif de production d'énergie selon l'une quelconque des revendications 1 à 9, dans lequel le moyen d'alimentation en gaz de combustible précité comprend de plus, comme partie de la séparation précitée, une seconde paroi (73; 93) dans laquelle la chambre de production d'énergie précitée (82) est séparée de la chambre de gaz de combustible précitée (72), agencée dans une direction perpendiculaire à la direction longitudinale précitée.

11. Dispositif de production d'énergie selon l'une quelconque des revendications 1 à 10, dans lequel le moyen de collecte de courant précité (75) comprend deux plaques de collecte de courant et les plaques sont agencées entre la plle de collecte précitée et la première paroi latérale précitée (74; 94) de la chambre de production d'énergie

précitée, parallèles à la direction longitudinale précitée.

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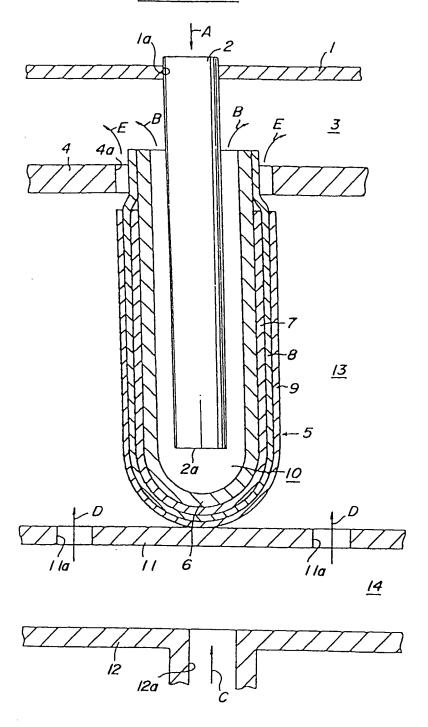
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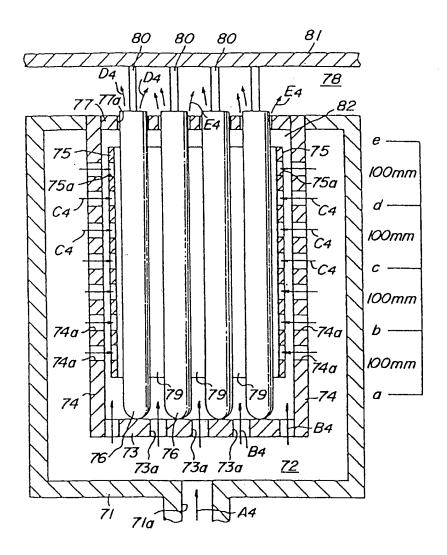
- 12. Dispositif de production d'énergie selon la revendication 13, dans lequel un certain nombre de trous traversants (75a) sont prévus dans les plaques de collecte de courant précitées (75).
- 13. Dispositif de production d'énergie selon l'une quelconque des revendications 1 à 12, dans lequel un certain nombre de trous traversants (73a, 74a) à travers lesquels passe le gaz de combustible dans la chambre de production d'énergie précitée, sont prévus dans au moins les premières parois précitées (74).
- 14. Dispositif de production d'énergie selon l'une quelconque des revendications 1 à 12, dans lequel au moins les premières parois précitées (93, 94) sont réalisées en un matériau poreux.
  - 15. Dispositif de production d'énergie selon la revendication 14, dans lequel au moins les premières parois précitées (74) sont réalisées en un matériau poreux formé en frittant une poudre métallique résistant à la réduction ou sont formées en un cermet poreux formé en frittant un mélange d'une poudre métallique résistant à la réduction et une poudre de céramique.
  - Dispositif de production d'énergie selon la revendication 15, dans lequel au moins les premières parois précitées
     (74) son réalisées en cermet nickel-zircone ou cermet cobalt-zircone.
  - 17. Dispositif de production d'énergie selon l'une quelconque des revendications 14 à 16, dans lequel au moins les premières parois précitées (74) ont une porosité dans la gamme de 10 à 90 %.
- 18. Dispositif de production d'énergie selon la revendication 17, dans lequel au moins les premières parois précitées (74) comprennent chacune un corps fritté réalisé en métal poreux ayant une porosité donnée et un certain nombre de régions conformées en ceinture formées de coulis agencés sur la surface du corps fritté à des espaces prédéterminés.
- 19. Equipement de production d'énergie selon la revendication 18, dans lequel la quantité de gaz de combustible transmis à travers les premières parois précitées (74) est contrôlée en variant lesdits espacements précités entre les coulis.

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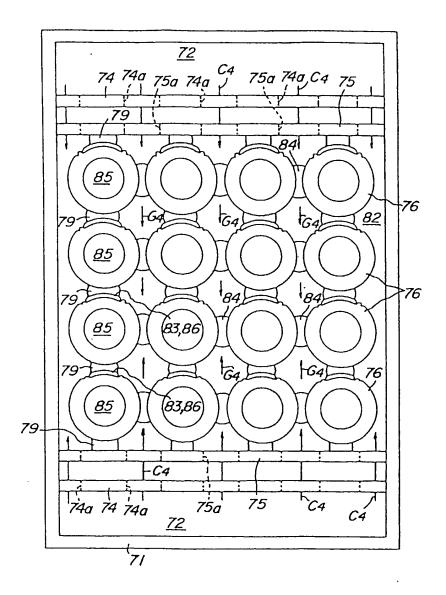
FIG\_I PRIOR ART

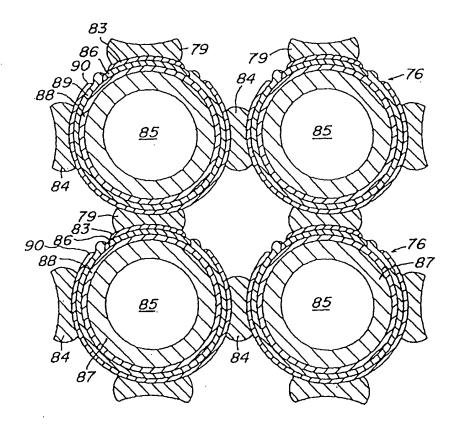


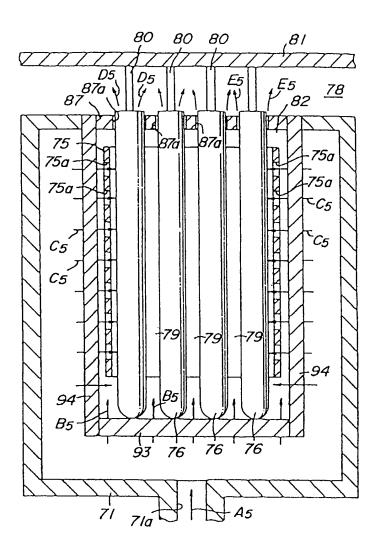
FIG\_2



FIG\_3







### F1G\_6

